

Relaxation rates of magnetization in $\text{LiYF}_4\text{:Ho}^{3+}$ crystals

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Abstract

Theory of magnetization relaxation in a diluted paramagnet based on calculations of relaxation rates due to electron–phonon coupling and magnetic dipole–dipole interactions is developed. Assuming fast establishment of equilibrium in the spin system and the Gaussian line shape of the spectral density of the dipole–dipole reservoir, we have derived a master equation for a paramagnetic ion. The developed approach is used to interpret dynamic magnetic measurements in $\text{LiYF}_4\text{:Ho}^{3+}$ single crystals. We show specific variations of low temperature magnetization dynamics at the magnetic field values corresponding to (anti-)crossings of electron–nuclear sublevels of the Ho^{3+} ground state. The measured dependence of the ac-susceptibility on frequency, holmium concentration and the applied magnetic field are described when taking into account the phonon bottleneck effect and the cross-relaxation. Evolution of the magnetization of the $\text{LiYF}_4\text{:Ho}^{3+}$ in the reversed field is considered.

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1. Introduction

Measurements of dynamic susceptibility [1], magnetization hysteresis in sweeping magnetic fields [2], and ^{19}F nuclear relaxation rates [3] in diluted paramagnets $\text{LiYF}_4\text{:Ho}^{3+}$ have revealed specific variations of the magnetization dynamics at the magnetic field values corresponding to crossings/anti-crossings of the electron–nuclear sublevels of the ground doublet state of Ho^{3+} ions in the crystal field of S_4 point symmetry. Results of these experimental investigations gave evidence for considerable changes (by orders of magnitude) of relaxation rates in the electron–nuclear system at the energy level (anti-)crossings. The goal of the present study is to clear up specific features of the dynamic susceptibility at liquid helium temperatures and the magnetization dynamics in sweeping magnetic fields. Different phenomena caused by the electron–phonon, magnetic dipole and hyperfine interactions are considered.

2. Kinetic equations for diluted paramagnets

The equation of motion for the density matrix $\rho(t)$ of a paramagnetic ion coupled to a phonon bath in the external magnetic

field in the basis of eigenfunctions of the Hamiltonian $H_0 + H_1$ (with eigenvalues E_n) of an electronic (or electron–nuclear) system can be written as follows [4]:

$$\frac{\partial \rho_n}{\partial t} = \sum_k W_{nk}^{\text{SL}} \rho_k, \quad \frac{\partial \rho_{nk}}{\partial t} = -(\gamma_{nk} + i\omega_{nk})\rho_{nk} \quad (n \neq k)$$

Here $\rho_n = \rho_{nn}$, H_0 is the Hamiltonian of a paramagnetic ion in the stationary magnetic field \mathbf{B} , $H_1(t)$ the energy of interaction with the external ac-field, γ_{nk} are the coherence decay rates, and the rate of variation of the external field is supposed to be much less than $\omega_{nk} = (E_n - E_k)/\hbar$. Off-diagonal elements of the relaxation matrix W_{kn}^{SL} ($n \neq k$) are transition probabilities induced by the electron–phonon interaction. For one-phonon transitions, $W_{nk}^{\text{SL}} = w_{nk}[(n(\omega_{nk}) + \Theta(\omega_{kn}))]$ ($n \neq k$); $W_{nn}^{\text{SL}} = -\sum_{k \neq n} W_{kn}^{\text{SL}}$, where w_{nk} is the probability of the spontaneous transition, $n(\omega_{nk})$ the phonon occupation number, Θ is the step function.

In the case of a finite concentration of paramagnetic ions, interactions between ions induce energy exchange (cross-relaxation), and the master Eq. (1) contains additional nonlinear terms [5,6]:

$$\dot{\rho}_n = \sum_m W_{nm}^{\text{SL}} \rho_m + R_n,$$

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